

1 **Supporting Information**

2

3 **Shipping Remains a Globally Significant Source of Anthropogenic PN**

4 **Emissions Even after 2020 Sulfur Regulation**

5 *Niina Kuittinen^a, Jukka-Pekka Jalkanen^b, Jenni Alanen^a, Leonidas Ntziachristos^a, Hanna*
6 *Hannuniemi^b, Lasse Johansson^b, Panu Karjalainen^a, Erkka Saukko^a, Mia Isotalo^a, Päivi Aakko-*
7 *Saksa^c, Kati Lehtoranta^c, Jorma Keskinen^a, Pauli Simonen^a, Sanna Saarikoski^b, Eija Asmi^b,*
8 *Tuomas Laurila^b, Risto Hillamo^b, Fanni Mylläri^a, Heikki Lihavainen^{b,d}, Hilkka Timonen^b, Topi*
9 *Rönkkö^a*

10 ^aAerosol Physics Laboratory, Physics Unit, Faculty of Engineering and Natural Sciences,
11 Tampere University, FI-33014, Tampere, Finland

12 ^bAtmospheric Composition Research, Finnish Meteorological Institute, P.O. Box 503, 00101
13 Helsinki, Finland

14 ^cVTT Technical Research Centre of Finland Ltd., P.O. Box 1000, 02044 VTT, Espoo, Finland

15 ^dSvalbard Integrated Arctic Earth Observing System, P.O. Box 156, 9171 Longyearbyen,
16 Norway

17

18

19 Number of pages: 16

20 Number of figures: 10

21 Number of tables: 7

22	Contents:
23	Supporting information text
24	
25	Figure S1. Dilution and positioning of devices in laboratory and on-board measurements.
26	Figure S2. Normalized particle number size distributions measured by EEPS.
27	Figure S3. Flight paths of the research aircraft during the chasing measurement of ships
28	operating in the Gulf of Finland.
29	Figure S4. Plot indicating the vectors needed to calculate time correction from the plume
30	sampling time frame to the real atmospheric time frame.
31	Figure S5. Evolution of geometric mean diameter and PN concentration over two in-plume
32	flights.
33	Figure S6. CPC and CO ₂ analyzer data for the plume chase measurements for the following
34	vessels: cargo 1983(a), cargo 2006(a), cargo 1983(b), cargo 2006(b), cargo 2000, cargo 2002,
35	cargo 2011, ferry 2008(a), and ferry 2008(b).
36	Figure S7. CPC and CO ₂ analyzer data for the plume chase measurements for the following
37	vessels: ferry 1993, ferry 1999, ferry 2000, ferry 2001, and ferry 2007.
38	Figure S8. CPC and CO ₂ analyzer data for the plume chase measurements for the following
39	vessels: ferry 2007, unidentified cargo, tanker 1980, tanker 2006, cargo 1982, cargo 2002, and
40	ferry 2008.
41	Figure S9. CPC and CO ₂ analyzer data for the plume chase measurements for the following
42	vessels: ferry 2008, ferry 1992, tanker 2004, and cargo 2008.
43	Figure S10. Global annual PN from shipping in 2016 and 2020, as well as the resulting PN from
44	hypothetical scenarios, where HFO would be mainly replaced by other fuel options than IFO.
45	
46	Table S1. Details of the test-bed engine used during laboratory testing.
47	Table S2. Properties of the fuels used in the laboratory and on-board measurements.
48	Table S3. Particle number emission factors used in the calculations. Standard deviation and
49	number of CPC data points are shown in parentheses.
50	Table S4. Fuel sulfur content (FSC) and engine after-treatment information for ships involved in
51	the chase study. Fuel sulfur content and exhaust after-treatment data has been provided by the
52	ship operators for part of the vessels. Standard deviation is given in parentheses.
53	Table S5. Marine fuel consumption during 2016 and comparison to previous studies.
54	Table S6. Global marine fuel consumptions (kg) used in calculations for the year 2020.
55	Table S7. Summary of methods applied in different parts of the study.

56 **Supporting Information Text**

57

58 **Applying condensation particle counters with varying cut-points.** In this work, CPCs with
59 different cut-points were applied in different phases of the study. In the first laboratory campaign
60 and on-board study, a CPC with 7nm cut-point was applied. In the second laboratory study, it was
61 expected that the particles resulted from the combustion of natural gas could appear in very small
62 size range and an ultrafine CPC with 3nm cut-point was chosen. In the plume chase studies, either
63 a CPC with 3nm or 10 nm cut-point was available. The effect of using a CPC with a cut-point of
64 10nm or 7nm compared to the CPC with 3nm cut-point can be estimated based on the particle
65 number size distributions for different fuels presented in this study (Figure 1A and 1B). For the
66 EF_{PN} presented in Figure 1C and applied to STEAM, the error introduced by using a CPC with
67 7nm cut-point compared to one with 3nm cut-point in the cases of HFO, IFO and BIO30 is
68 negligible, with less than 0.05% of particles being uncounted due to the higher cut-point. In the
69 plume chase measurements, all the ships whose fuel information could be obtained burned either
70 HFO or IFO, and it is assumed that the error introduced by the 10nm cut-point in part of the
71 measurements is also small, between 0.05 and 0.3%. However, if LNG ships would have been
72 chased, the influence could have been more significant, up to 4%.

73 **Calculating the plume age.** During the Skyvan flights, the maximum in-plume flight duration,
74 on which continuous size distribution spectra, particle number concentration data, and gas
75 component concentrations could be obtained, was 120 s. As the velocity of the aircraft was much
76 higher than the prevailing wind and ship velocities, an effective aging of the plume of up to 600 s
77 was captured. The proper age of the exhaust during Skyvan measurements was calculated from
78 the velocities of the ship, aircraft and the prevailing wind. The time correction from the sampling

79 timeframe to the real atmospheric ageing timeframe has been calculated analogously to general
80 Doppler Effect (1) (see Supporting Information and Figure S7)

$$81 \quad \gamma = \frac{\|V_a - W\|}{\|V_s - W\|} \quad (1)$$

82 where V_a , W , and V_s denote the aircraft, wind and ship velocity vectors along the plume direction,
83 respectively, with the V_a required to be for an in-plume flight. The wind speed and direction were
84 obtained as an average of three nearby weather stations at Helsinki (Helsingin majakka), Porvoo
85 (Kalbådagrund) and Kirkkonummi Mäkiluoto. The corresponding wind data was then calculated
86 at the aircraft flight height using the power-law wind velocity profile method (2) and an assumption
87 of neutral atmospheric conditions.

88 The age of the ship plume when it crosses the sampling inlet in the aircraft can be resolved
89 analogous to Doppler phenomenon in the general case with a moving source and a moving
90 observer. In the most simple case with wind speed (W) zero, the ageing factor would naturally be
91 the ratio of aircraft speed (V_a) and ship speed (V_s). In the most complex case with different wind,
92 aircraft and ship speed directions, the angles between the three need to be taken into account (see
93 Figure S7). In a frame of reference moving with air, the plume length emitted in time $\Delta\tau$ can be
94 expressed as $\Delta S_s = \Delta\tau(W - V_s)$ and plume length captured by the aircraft as $\Delta S_a = \Delta\tau(W - V_a)$.
95 Therefore, time dilation, i.e. the time correction γ from the sampling timeframe to the real
96 atmospheric ageing timeframe can be given as

$$97 \quad \gamma = \frac{|\Delta S_a|}{|\Delta S_s|} = \frac{|(V_a - W)|}{|(V_s - W)|}.$$

98 The situation is analogous to Doppler formula in the general case. See the example of Neipp et al.
99 (1), equation (8). The time correction from the sampling timeframe to the real atmospheric ageing
100 timeframe is defined as the age of the emission divided by time measured. Thus,

101 $\gamma = 1/\omega / 1/\omega' = \omega'/\omega = (\mathbf{c}-\mathbf{v}_O) \cdot \mathbf{n} / (\mathbf{c}-\mathbf{v}_O) \cdot \mathbf{n}.$

102 Because here the aircraft was flying in the plume, $\gamma = |(\mathbf{v}_O-\mathbf{c})| / |(\mathbf{v}_S-\mathbf{c})|$. If we denote $\mathbf{c} = \mathbf{W}$, $\mathbf{v}_O =$
103 \mathbf{V}_a , and $\mathbf{v}_S = \mathbf{V}_s$ it follows that $\gamma = |(\mathbf{V}_a-\mathbf{W})| / |(\mathbf{V}_s-\mathbf{W})|$.

104 In calculating the plume age, flight height was considered only in the wind speed profile. However,
105 the time it takes for the plume to rise to the flying height was not accounted for, and thus, the actual
106 ageing times of the plumes may be longer than the calculated values. Other sources of uncertainty
107 are the wind speeds and directions; the wind velocity at the measurement location was assumed to
108 be the same as the one at the nearest weather station corrected with the power-law wind velocity
109 profile method for the height difference of the weather station and the aircraft. In addition, the ship
110 trajectory was assumed straight.

111 **Inclusion of scrubbers in STEAM model.** For the base year 2016, scrubber installations were
112 found in 210 vessels and the installation dates and equipment types (closed/open loop, hybrid)
113 were included in STEAM and obtained from shipyard reference lists, equipment manufacturers,
114 and ship owner fleet descriptions. According to classification society guidance, 3% increase in
115 power and fuel consumption due to pumps and coolers was implemented for vessels using open-
116 loop scrubbers (3). While accounting for added power consumption due to scrubbers, in the run
117 for 2016, we did not include the effect of scrubber on EF_{PN} , since only about 0.2% of the fleet
118 (1.4% of global fuel consumption) was equipped with scrubber, and the impact of this assumption
119 was expected to be negligible. The scrubber reduction efficiency was determined from on-board
120 measurements. Due to the fairly low sulfur level of the HFO burned by the ship (0.7%S), the
121 scrubber reduction efficiency was applied to the EF_{PN} for 2.2%S HFO and this was used in the
122 global scenario calculations to represent ships equipped with scrubbers burning HFO.

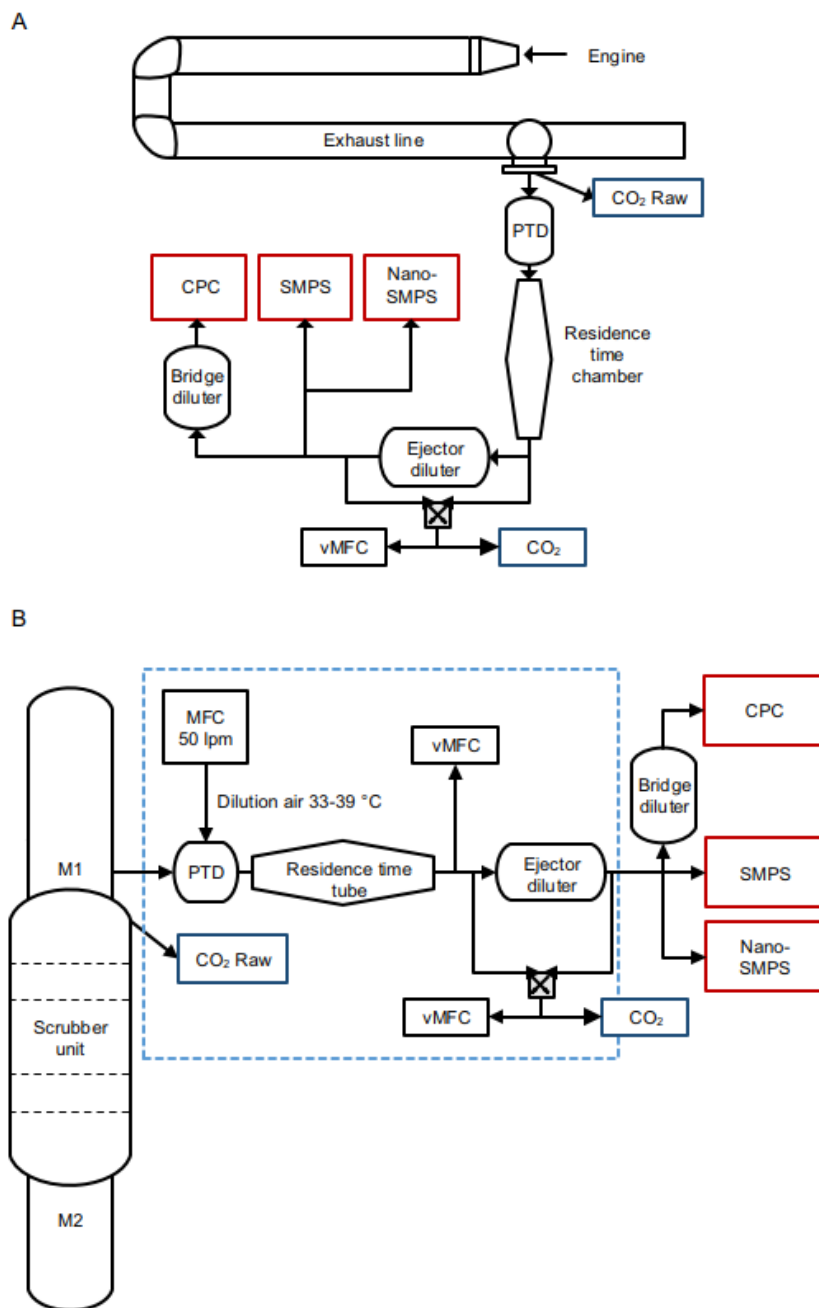
123 **References**

124 1. C. Neipp *et al.* An analysis of the classical Doppler effect. *Eur. J. Phys.* **24**, 407–505
125 (2003).

126 2. J. H. Seinfeld and S. N. Pandis. *Atmospheric Chemistry and Physics: From Air Pollution*
127 *to Climate Change*. (John Wiley & Sons, Hoboken, ed. 3, 2016) p. 700.

128 3. “Exhaust Gas Scrubber Systems” (American Bureau of Shipping, 2018);
129 [https://ww2.eagle.org/content/dam/eagle/advisories-and-debriefs/exhaust-gas-scrubber-systems-
advisory.pdf](https://ww2.eagle.org/content/dam/eagle/advisories-and-debriefs/exhaust-gas-scrubber-systems-
130 advisory.pdf)

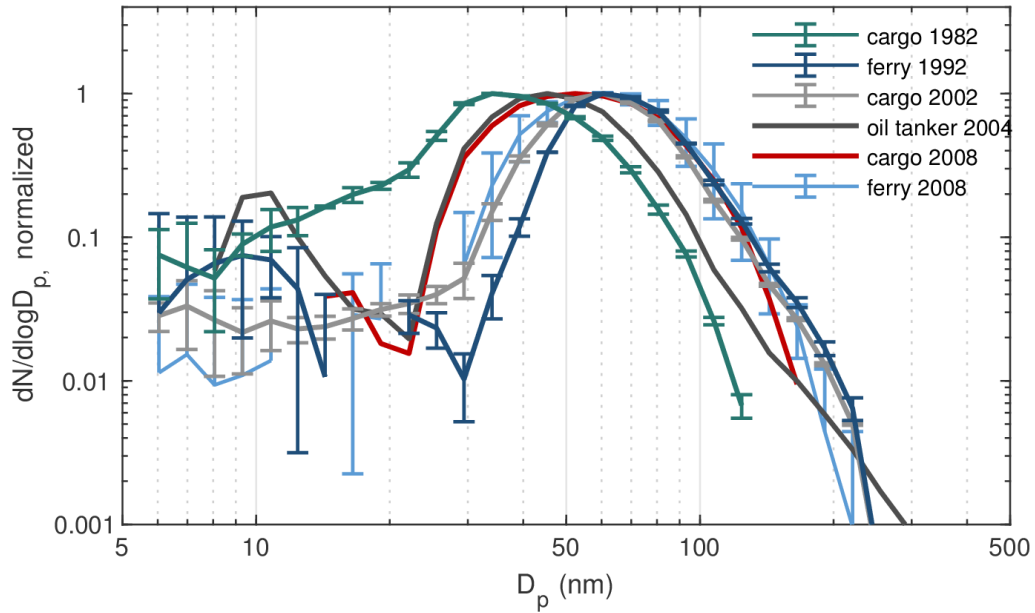
131



133

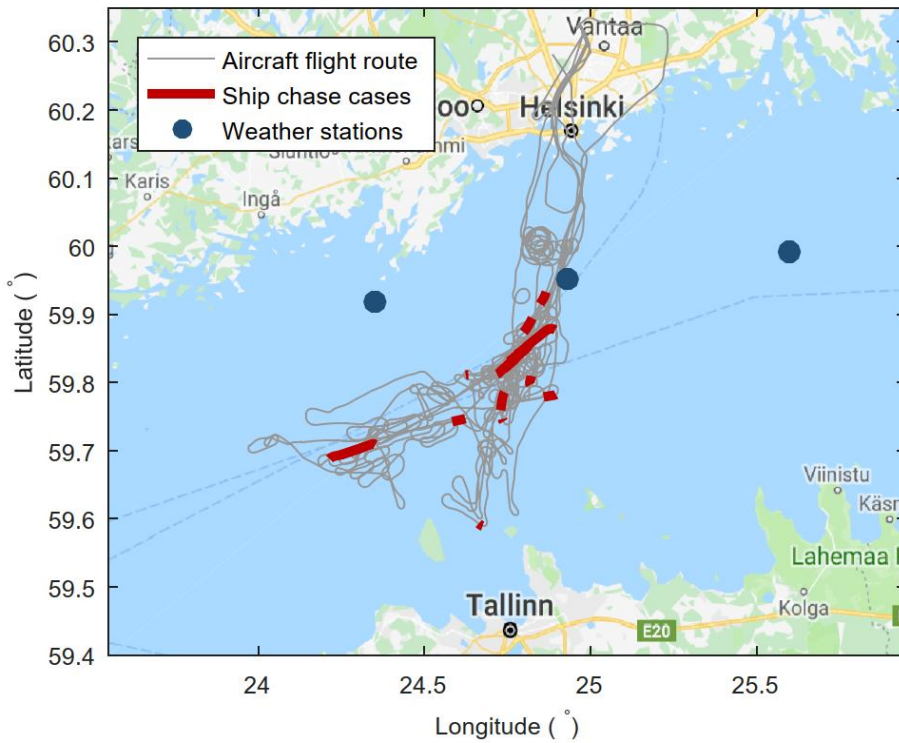
134 **Figure S1.** Dilution and positioning of devices in laboratory and on-board measurements. A.
 135 Measurement setup during of test-bench measurements. Exhaust was sampled from 5.9 m
 136 downstream of the turbo charger. B. Measurement devices and sampling locations during on-
 137 board testing where exhaust was sampled right before and after the scrubber. M1 marks sampling
 138 location in the exhaust tunnel after scrubber and M2 sampling location right before the scrubber
 139 unit.

140

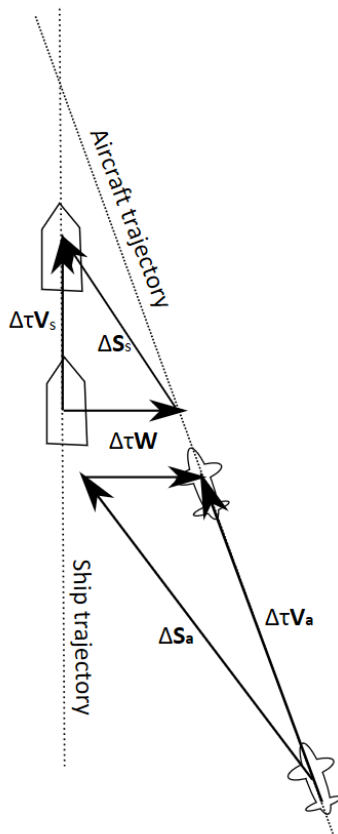


141
 142 **Figure S2.** Normalized particle number size distributions measured by EEPS as a mean over in-
 143 plume flight for all the ships for which EEPS measurement was available. Error bars denote one
 144 standard deviation when multiple chases were conducted.

145



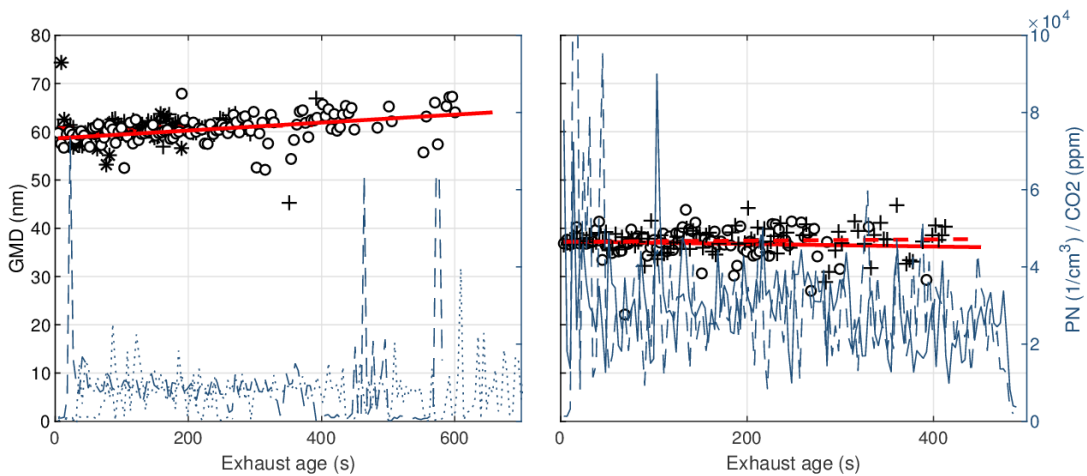
146
 147 **Figure S3.** Flight paths of the research aircraft during the chasing measurement of ships
 148 operating in the Gulf of Finland.



149

150 **Figure S4.** Plot indicating the vectors needed to calculate time correction from the plume
 151 sampling time frame to the real atmospheric time frame.

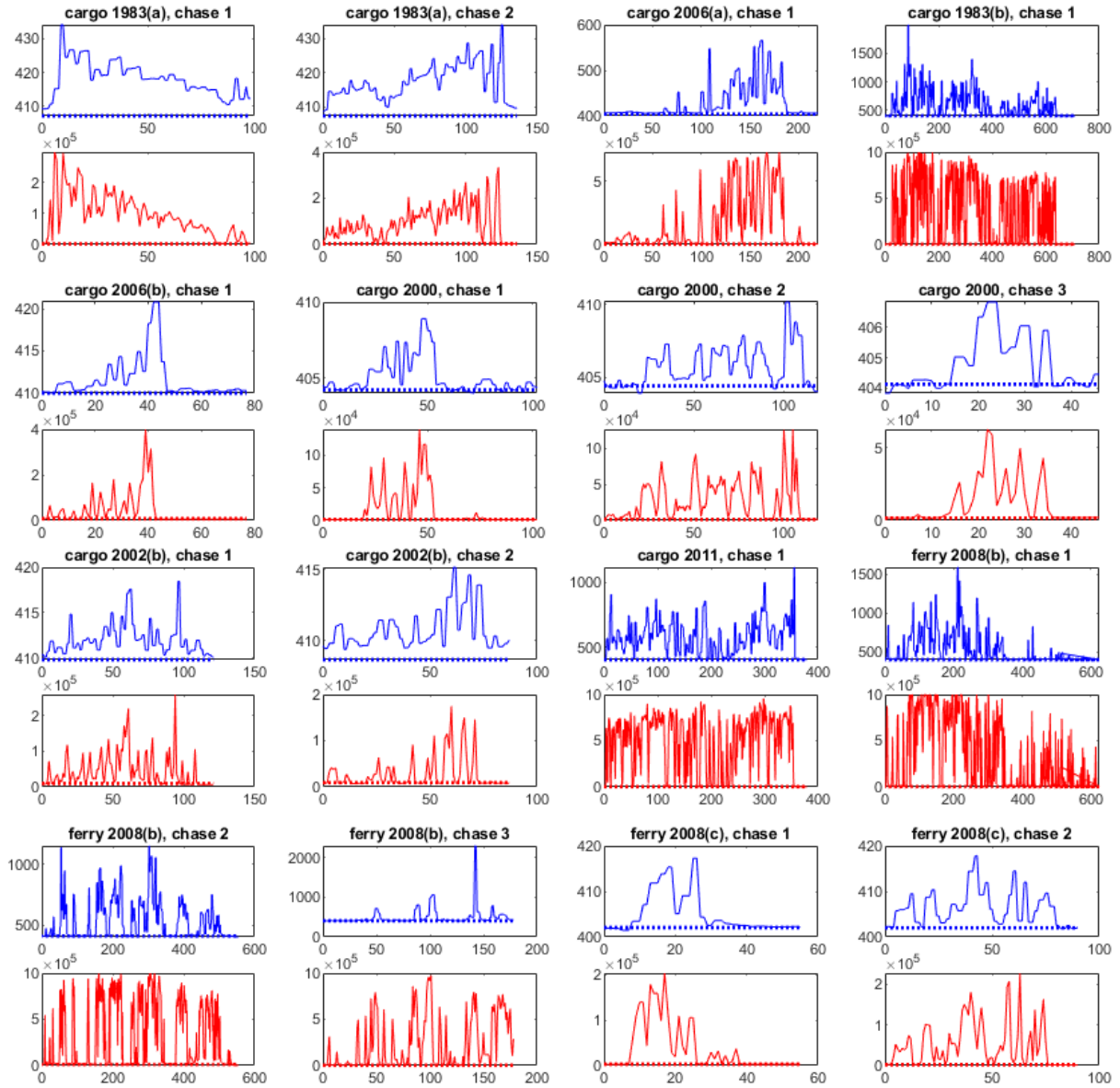
152



153

154 **Figure S5.** Evolution of geometric mean diameter and PN concentration over two in-plume
 155 flights shown in Figure 2. Here particle number concentrations are shown as normalized by CO2
 156 concentration. Varying symbols correspond to separate approaches.

157

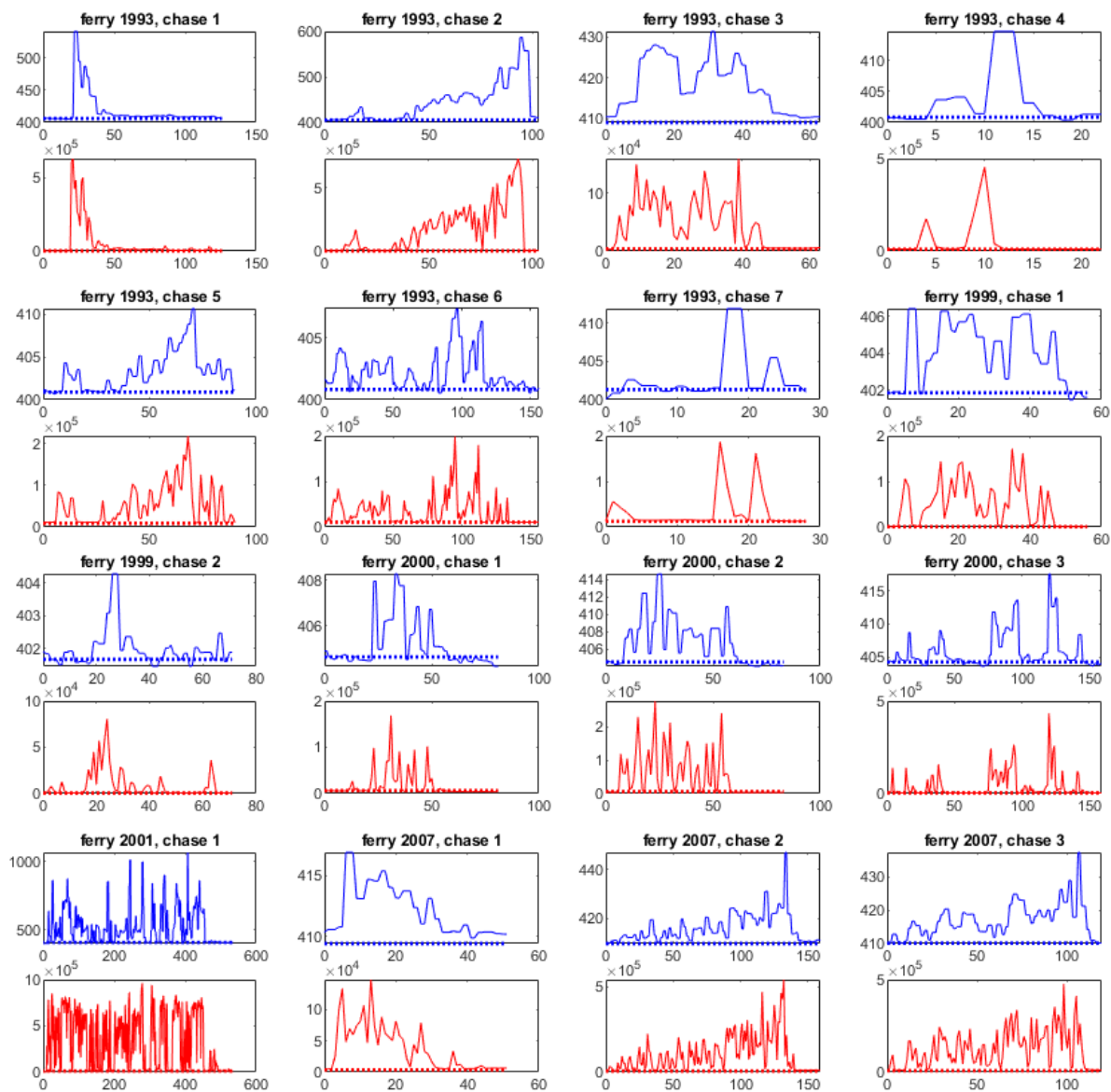


158

159 **Figure S6.** CPC and CO₂ analyzer data for the plume chase measurements for the following
 160 vessels: cargo 1983(a), cargo 2006(a), cargo 1983(b), cargo 2006(b), cargo 2000, cargo 2002,
 161 cargo 2011, ferry 2008(a), and ferry 2008(b). Solid blue lines mark CO₂ concentrations (ppm)
 162 during the approaches and dotted blue lines mark the average background CO₂ concentrations
 163 (ppm). Analogously, solid red lines mark PN concentrations (1/cm³) during the approaches and
 164 dotted red lines mark the average background PN concentrations (1/cm³). All concentrations are
 165 plotted against elapsed time (s).

166

167

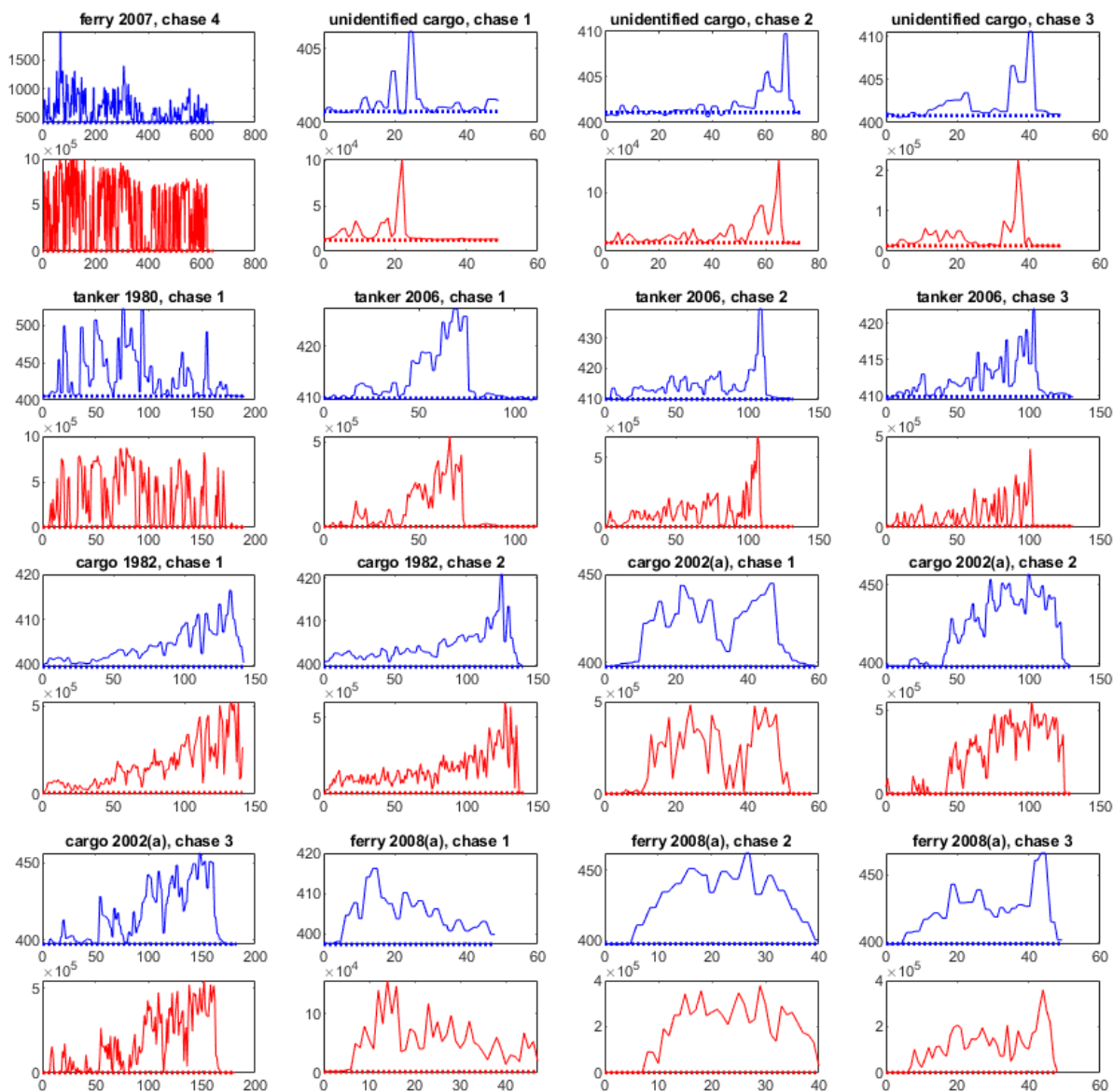


168

169 **Figure S7.** CPC and CO₂ analyzer data for the plume chase measurements for the following
 170 vessels: ferry 1993, ferry 1999, ferry 2000, ferry 2001, and ferry 2007. Solid blue lines mark
 171 CO₂ concentrations (ppm) during the approaches and dotted blue lines mark the average
 172 background CO₂ concentrations (ppm). Analogously, solid red lines mark PN concentrations
 173 (1/cm³) during the approaches and dotted red lines mark the average background PN
 174 concentrations (1/cm³). All concentrations are plotted against elapsed time (s).

175

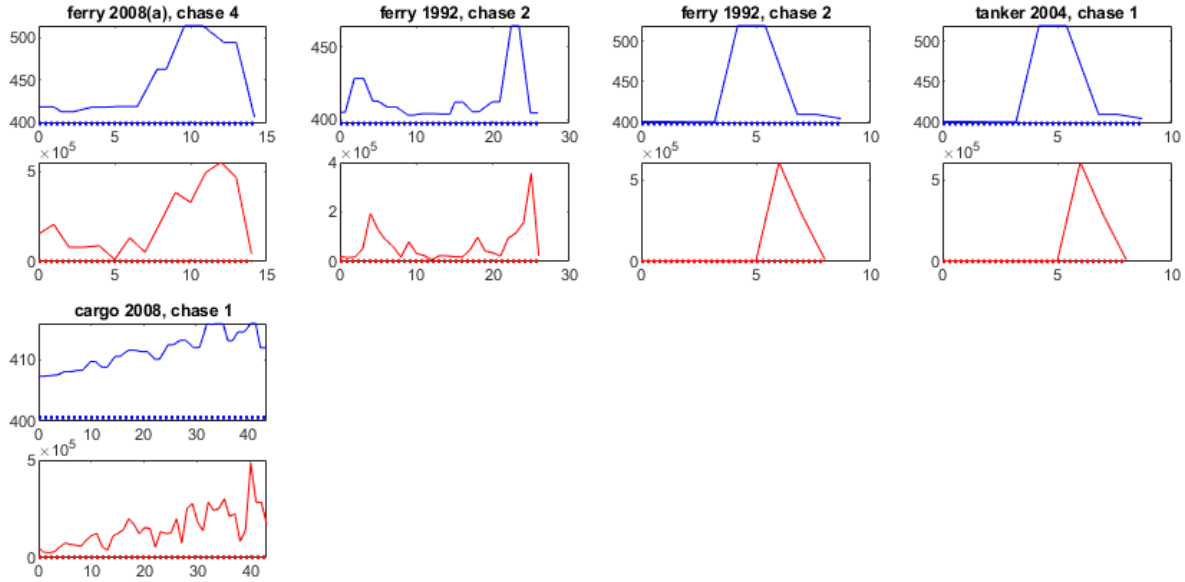
176



177

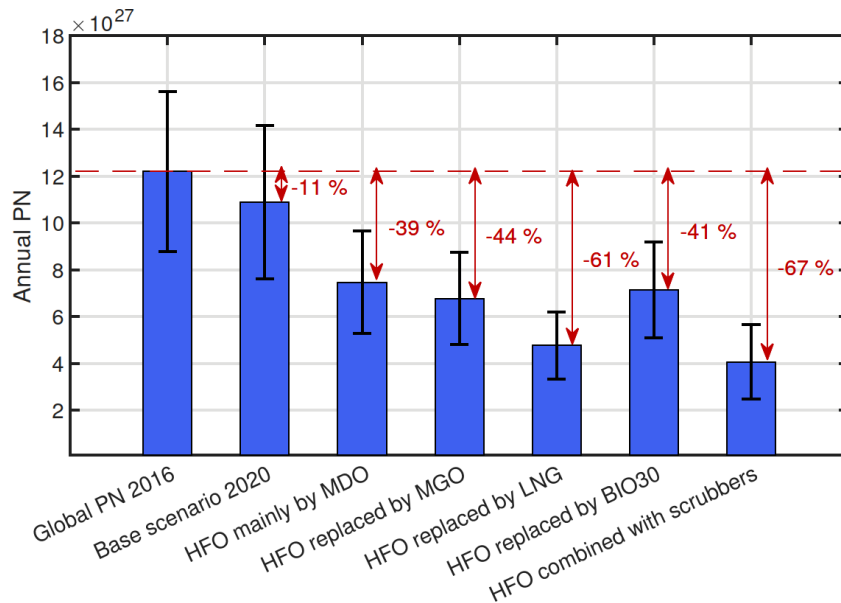
178 **Figure S8.** CPC and CO₂ analyzer data for the plume chase measurements for the following
 179 vessels: ferry 2007, unidentified cargo, tanker 1980, tanker 2006, cargo 1982, cargo 2002, and
 180 ferry 2008. Solid blue lines mark CO₂ concentrations (ppm) during the approaches and dotted
 181 blue lines mark the average background CO₂ concentrations (ppm). Analogously, solid red lines
 182 mark PN concentrations (1/cm³) during the approaches and dotted red lines mark the average
 183 background PN concentrations (1/cm³). All concentrations are plotted against elapsed time (s).

184



185

186 **Figure S9.** CPC and CO₂ analyzer data for the plume chase measurements for the following
 187 vessels: ferry 2008, ferry 1992, tanker 2004, and cargo 2008. Solid blue lines mark CO₂
 188 concentrations (ppm) during the approaches and dotted blue lines mark the average background
 189 CO₂ concentrations (ppm). Analogously, solid red lines mark PN concentrations (1/cm³) during
 190 the approaches and dotted red lines mark the average background PN concentrations (1/cm³). All
 191 concentrations are plotted against elapsed time (s).



192

193 **Figure S10.** Global annual PN from shipping in 2016 and 2020, as well as the resulting PN from
 194 extreme scenarios, where HFO would be mainly replaced by other fuel options than IFO. Error
 195 bars denote the combined uncertainty in the estimates. The red annotations mark potential
 196 reductions in PN achieved by the presented scenarios.

197 **Table S1.** Details of the test-bed engine used during laboratory testing.

Wärtsilä Vasa 4R32 LN E	Liquid fuel operation mode	Natural gas operation mode*
Nominal power (kW)	1640	1400
Number of cylinders	4	4
Speed (rpm)	750	750
Bore (mm)	320	320
Stroke (mm)	350	350
Compression ratio	13.8	12
Rotating direction	clockwise	clockwise
Firing order	1-3-4-2	1-3-4-2
Exhaust valve opens / closes	56° bbdc / 44° atdc	34,8° bbdc / 7,1° atdc
Inlet valve opens / closes	52° bt dc / 28° bbdc	9,1° bt dc / 35,3° bbdc
Mechanical injection system	Pump L'Orange PEO-GO52, valve L'Orange VUO-U156, nozzle Wärtsilä part number 009422407	Pump L'Orange PEO-GO52, valve L'Orange VUO-G259b, nozzle VUO-U9121b/E / PA AE010974
Static injection advance	12.3°	1: 16.5°, 2: 17.0°, 3: 17.0°, 4: 17.0°
Turbocharger	BBC VTR 254	ABB TPS52-F33 HT 481009

* The modification of the engine to NG operation included, for instance, installing gas admission valves on the cylinder heads, replacing cylinder heads as well as liners, pistons, camshaft and turbocharger. In addition, engine control system was updated and a pilot fuel line with a high pressure pilot fuel pump was added. Exhaust gas piping to stack was equipped with burst disk for safety.

198

199 **Table S2.** Properties of the fuels used in the laboratory and on-board measurements.

200

Fuel parameter	MGO *	MDO	IFO	HFO 2.2%S	BIO30	HFO 0.7%S
Density (kg/m ³)	838 / 835 (15 ° C)	879 (15 ° C)	906 (40 ° C)	979 (40 ° C)	866 (40 ° C)	873 (15 ° C)
Viscosity (mm ² /s)	3.3 / 2.6 (40 ° C)	4.1 (40 ° C)	127 (50 ° C)	187 (80 ° C)	6.7 (40 ° C)	886 (40 ° C)
Sulphur (mg/kg)	6.3 / 5.9	822	3 750	22 200	< 5	6520
Carbon (wt-%)	86.1 / 86.2	87.4	87.2	85.5	83.5	87.7
Hydrogen (wt-%)	13.9 / 13.8	13.9	12.6	10.3	13.2	11.5
Oxygen (wt-%)	< 0.5 / 0.5	< 0.5	< 0.5	0.5	3.9	0.6
Nitrogen (mg/kg)	19.3 / 61.8	367	2 275	5408	28.5	2490
Ash (wt-%)	< 0.005	< 0.005	0.038 (775 ° C)	0.094 (775 ° C)	< 0.005	< 0.005 (775 ° C)
Heating value, lower (kJ/kg)	42.7 / 42.9	42.2	42.1	40.3	40.7	40.8

* MGO was also used as the pilot fuel in MDO and NG measurements, two batches of fuel were tested

201

202 **Table S3.** Particle number emission factors used in the calculations. Standard deviations are
 203 shown in parentheses.

Fuel	Cruising		Maneuvering		Weighted average	
	EF _{PN} (#/kg _{fuel})	EF _{PN} (#/kWh)	EF _{PN} (#/kg _{fuel})	EF _{PN} (#/kWh)	EF _{PN} (#/kg _{fuel})	EF _{PN} (#/kWh)
NG*	1.38 x 10 ¹⁶ (± 2.36 x 10 ¹⁵)	2.35 x 10 ¹⁵ (± 4.03 x 10 ¹⁴)	2.11 x 10 ¹⁶ (± 1.34 x 10 ¹⁵)	4.29 x 10 ¹⁵ (± 2.72 x 10 ¹⁴)	1.60 x 10 ¹⁶	2.93 x 10 ¹⁵
MGO	2.37 x 10 ¹⁶ (± 2.04 x 10 ¹⁵)	4.81 x 10 ¹⁵ (± 4.13 x 10 ¹⁴)	3.26 x 10 ¹⁶ (± 1.63 x 10 ¹⁵)	7.32 x 10 ¹⁵ (± 3.67 x 10 ¹⁴)	2.64 x 10 ¹⁶	5.34 x 10 ¹⁵
MDO	2.83 x 10 ¹⁶ (± 2.92 x 10 ¹⁵)	5.76 x 10 ¹⁵ (± 5.95 x 10 ¹⁴)	3.23 x 10 ¹⁶ (± 2.60 x 10 ¹⁵)	7.11 x 10 ¹⁵ (± 5.72 x 10 ¹⁴)	2.95 x 10 ¹⁶	6.17 x 10 ¹⁵
IFO	4.37 x 10 ¹⁶ (7.29 ± x 10 ¹⁵)	8.88 x 10 ¹⁵ (± 1.48 x 10 ¹⁵)	5.69 x 10 ¹⁶ (± 2.50 x 10 ¹⁵)	1.36 x 10 ¹⁶ (± 5.99 x 10 ¹⁴)	4.76 x 10 ¹⁶	1.03 x 10 ¹⁶
HFO 2.2%S	5.39 x 10 ¹⁶ (± 2.77 x 10 ¹⁵)	1.16 x 10 ¹⁶ (± 5.94 x 10 ¹⁴)	5.83 x 10 ¹⁶ (± 9.42 x 10 ¹⁴)	1.47 x 10 ¹⁶ (± 2.37 x 10 ¹⁵)	5.52 x 10 ¹⁶	1.26 x 10 ¹⁶
BIO30	2.28 x 10 ¹⁶ (± 1.24 x 10 ¹⁵)	4.73 x 10 ¹⁵ (± 2.58 x 10 ¹⁴)	3.44 x 10 ¹⁶ (± 1.02 x 10 ¹⁵)	8.24 x 10 ¹⁵ (± 2.44 x 10 ¹⁴)	2.63 x 10 ¹⁶	5.78 x 10 ¹⁵
HFO 0.7% S, Engine A						
Before scrubber	3.20 x 10 ¹⁶ (± 1.08 x 10 ¹⁵)	6.28 x 10 ¹⁵ (± 2.12 x 10 ¹⁴)	4.24 x 10 ¹⁶ (± 6.78 x 10 ¹⁴)	8.47 x 10 ¹⁵ (± 1.35 x 10 ¹⁴)	3.52 x 10 ¹⁶	6.93 x 10 ¹⁵
After scrubber	2.73 x 10 ¹⁵ (± 5.77 x 10 ¹³)	5.36 x 10 ¹⁴ (± 1.13 x 10 ¹³)	4.28 x 10 ¹⁵ (± 4.27 x 10 ¹⁴)	8.53 x 10 ¹⁴ (± 8.51 x 10 ¹³)	3.20 x 10 ¹⁵	6.31 x 10 ¹⁴
HFO 0.7%S, Engine B						
Before scrubber	2.50 x 10 ¹⁶ (± 1.05 x 10 ¹⁵)	4.83 x 10 ¹⁵ (± 2.02 x 10 ¹⁴)	3.20 x 10 ¹⁶ (± 9.32 x 10 ¹⁴)	6.51 x 10 ¹⁵ (± 1.89 x 10 ¹⁴)	2.71 x 10 ¹⁶	5.33 x 10 ¹⁵
After scrubber	7.72 x 10 ¹⁵ (± 5.02 x 10 ¹⁴)	1.49 x 10 ¹⁵ (± 9.70 x 10 ¹³)	n/a	n/a	n/a	n/a

*NG burned in dual-fuel operation with MGO as pilot fuel

205 **Table S4.** Fuel sulphur content (FSC) and engine after-treatment information for ships involved
 206 in the chase study. Fuel sulphur content and exhaust after-treatment data has been provided by
 207 the ship operators for part of the vessels. Standard deviation is given in parentheses.

Vessel	EF _{PN} (1/kg _{fuel})	Number of chases	Fuel	FSC (wt%)	Exhaust after-treatment	Classification
cargo 1982	5.8 x 10 ¹⁶ (±4.9 x 10 ¹⁵)	3	HFO	0.99	none	Container Ship
ferry 1992	5.6 x 10 ¹⁵	2	HFO	0.5	none	Ro-Ro/Passenger Ship
cargo 2002(a)	1.5 x 10 ¹⁶	2	HFO	0.97	none	Ro-Ro Cargo
tanker 2004	2.3 x 10 ¹⁶	1	n/a	n/a	n/a	Oil Products Tanker
cargo 2008	6.9 x 10 ¹⁵	1	HFO	0.94	none	Container Ship
ferry 2008(a)	9.0 x 10 ¹⁵ (±1.7 x 10 ¹⁵)	4	HFO	1	none	Ro-Ro/Passenger Ship
ferry 2008(b)	4.2 x 10 ¹⁵ (±6.0 x 10 ¹³)	3	IFO	0.87	none	Ro-Ro/Passenger Ship
ferry 2001	5.0 x 10 ¹⁵	1	n/a	n/a	n/a	Ro-Ro/Passenger Ship
ferry 2007	2.0 x 10 ¹⁶ (±1.0 x 10 ¹⁶)	4	HFO	1	none	Passenger Ship
cargo 2006(a)	9.6 x 10 ¹⁵	1	n/a	n/a	n/a	Container Ship
tanker 1980	1.8 x 10 ¹⁶	1	n/a	n/a	n/a	Oil/Chemical Tanker
cargo 2011	4.7 x 10 ¹⁵	1	HFO	0.97	none	Ro-Ro Cargo
ferry 1993	1.9 x 10 ¹⁶ (±1.0 x 10 ¹⁶)	7	HFO	1	SCR	Ro-Ro/Passenger Ship
cargo 1983(a)	1.5 x 10 ¹⁶	2	n/a	n/a	n/a	Container Ship
tanker 2006	3.9 x 10 ¹⁶ (±1.6 x 10 ¹⁵)	3	n/a	n/a	n/a	Crude Oil Tanker
cargo 2006(b)	3.4 x 10 ¹⁶	1	n/a	n/a	n/a	Container Ship
cargo 2002(b)	2.1 x 10 ¹⁶	2	n/a	n/a	n/a	Container Ship
unidentified cargo	2.5 x 10 ¹⁶ (±3.3 x 10 ¹⁵)	3	n/a	n/a	n/a	n/a
ferry 2000	3.5 x 10 ¹⁶ (±1.7 x 10 ¹⁵)	3	HFO	0.97	none	Ro-Ro/Passenger Ship
ferry 2008(c)	1.9 x 10 ¹⁶	2	HFO	0.5	none	Ro-Ro/Passenger Ship
ferry 1999	4.1 x 10 ¹⁶	2	HFO	0.97	none	Ro-Ro/Passenger
cargo 1983(b)	2.9 x 10 ¹⁵	1	n/a	n/a	n/a	Reefer
cargo 2000	3.0 x 10 ¹⁶ (±3.4 x 10 ¹⁵)	3	n/a	n/a	n/a	General Cargo

Fuel, fuel sulfur content (FSC), and exhaust after-treatment information has been provided by the shipping companies for part of the ships. Ships are labeled according to their classification in Marine Traffic database (<https://marinetraffic.com>). During the measurements, FSC in Baltic Sea SECA was limited to 1.0% S.

208

209

210

211 **Table S5.** Marine fuel consumption during 2016 and comparison to previous studies.

Fuel	Annual consumption (million tons) ^a	Contribution to total (%)	Consumption with inland contribution (million tons)	IEA, 2011	STEAM, 2015	GHG3, 2012
Residual (HFO)	185	72.1	188	191	195	
Diesel (MDO)	44	17.1	48	62 ^b	81 ^b	
Gasoil (MGO)	23	9.0	27			
LNG	5	1.9	5			
Total	257		267	254	276	300

^a Excluding rivers, lakes

^b Sum of MDO, MGO and LNG

212

213

214 **Table S6.** Global marine fuel consumptions (Mt) used in calculations for the year 2020.

215

Fuel consumptions (Mt)	LNG	MGO	MDO	HFO + scrubber	IFO	BIO30
Base scenario 2020	8.20	24.5	46.8	10.9	180	2.73
HFO mainly replaced by MDO	8.20	24.5	2.24	10.9	0	2.73
HFO mainly replaced by MGO	8.20	202	4.68	10.9	0	2.73
HFO mainly replaced by LNG	161	24.5	4.68	10.9	0	2.73
HFO mainly replaced by BIO30	8.20	24.5	4.68	10.9	0	189
HFO mainly replaced by scrubbers	8.20	24.5	4.68	199	0	2.73

216

217 **Table S7.** Summary of methods applied in different parts of the study.

Study	Method	Short description	Reference
Laboratory and on-board tests	Dilution of the sample	Primary dilution: PTD + residence time tube + ejector dilution to mimic ambient dilution conditions	(1, 36-38)
		Secondary dilution: Ejector diluters and bridge type diluters	(39)
	Particle number concentration	Airmodus A20 Condensation Particle Counter, cut-point 7nm	
	Particle number size distribution	SMPS: DMA 3071A and CPC 3775, TSI Inc., size range 10-414 nm; nano-SMPS: DMA 3085 and CPC 3776, TSI Inc., size range 3-64 nm	
	CO ₂ concentration	SIDOR Sick Maihak gas analyzer	
Ship plume measurements	Sample intake	Skyvan aircraft: BMI Isokinetic Inlet System (Model 1200, Brechtel Manufacturing Inc., USA); two lighter aircrafts: air intake tube with OD of 10 mm and length of 2 m	
	Particle number concentration	Skyvan aircraft: TSI Inc. 3010 Condensation Particle Counter, cut-point 10nm; two lighter aircrafts: TSI Inc. 3776 Condensation Particle Counter, cut-point 3 nm	
	Particle number size distribution	Skyvan aircraft: TSI Inc. Engine Exhaust Particle Sizer, size range 5.6-560 nm (default inversion algorithm)	(41)
	CO ₂ concentration	Picarro G1301-m	(40)

	Calculation of PN emission factors	PN emission rate calculation based on time integrals of CPC and CO ₂ analyzer signal peaks. Emission factor calculation based on PN emission rate and emission factor of 3.107 kg/kgfuel for CO ₂ .	(42 eq. 1) (42 eq. 2b, 43)
	Aircraft position, speed, and flight height	GPS recording	
	Ship position and speed	GPS recording	
	Wind speed and direction	Average of three nearby weather stations at Helsinki (Helsingin majakka), Porvoo (Kalbådagrund) and Kirkkonummi Mäkiluoto Corresponding wind data at the aircraft flight height calculated using the power-law wind velocity profile method and assumption of neutral atmospheric conditions.	(SI Ref. 2)
	Plume age	Method analogous to general Doppler effect described in the SI	SI, (SI Ref. 1)
Modelling global PN from ship traffic	Calculating global fuel consumption by fuel type and spatial distribution of PN emissions	STEAM Ship Traffic Emission Assessment Model (Version 3)	(44-47)
	Ship positions and speeds	Global Automatic Identification System (AIS) data, Orbcomm	
	Technical descriptions of ships	IHS Markit database	
	Calculation of fuel usage	Method based on water resistance calculated based on AIS data, separately for main and auxiliary engines Hollenbach water resistance method has been described in Jalkanen et al. (2012) and references within. In short, the method involves the evaluation of wet surface area of the hull, propulsion efficiency determination, hull form calculations and Reynolds/Froude number determination. ITTC friction formula is applied.	(45, and references within)
	Mitigating incomplete AIS data	Smart routing method was used to fill in gaps in AIS. This interpolation routine applied for vessel location determination considers potential land masses between the known two locations.	(47, Appendix B)
	Designation of vessel specific fuel type	Designation based on technical description of engine characteristics and vessel position (following SECA restrictions)	
	Determining specific fuel oil consumption (SFOC) for an engine	Method assuming parabolic dependency of SFOC as function of engine load, operating on the engine manufacturer data of SFOC for each engine	(48)
	Considering effect of scrubbers on SFOC	Method applying 3% additional power consumption for ships with open-loop scrubbers	SI, (SI Ref. 3)
	Considering uncertainty of the global fuel consumption results	Comparisons to vessel fuel reports (Jalkanen et al. 2009, Jalkanen et al. 2012) as well as comparisons to EU MRV reporting scheme.	(44,45)
Determining share of different fuel options in 2020	Estimates as share of global fuel usage were derived from industry reports.	(49,50)	

218

219